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COMPOSITION OF THE NATURAL GAS USED IN TWENTY-FIVE CITIES

WITH A DISCUSSION OF THE PROPERTIES OF NATURAL GAS

BY

G. A. BURRELL

AND

G. G. OBERFELL

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COMPOSITION OF THE NATURAL GAS USED IN TWENTY-FIVE CITIES; WITH A DISCUSSION OF THE PROPERTIES OF NATURAL GAS.

By G. A. Burrell and G. G. Oberfell.

INTRODUCTION.

In the course of its investigations looking to a more efficient utilization of the fuel resources in the United States, the Bureau of Mines has collected and analyzed samples of natural gas from wells in many oil and gas fields. This report presents the results of an examination of natural gas as supplied to 25 cities. No such comparison has hitherto been made, and the results are of interest because they show how the gas in one region differs from that in another and emphasize the points of similarity.

It is generally assumed that methane is the predominant constituent of natural gas, and that the heating value of natural gas is approximately 1,000 B. t. u. per cubic foot at 0° C. (32° F.) and 760 mm. pressure. One object of this investigation was to test the natural gas used in many cities in order to see how closely these assumptions approach the truth.

SAMPLING METHODS USED.

It was impossible for the representatives of the Bureau of Mines to personally visit the cities mentioned in this report and collect samples, hence material and instructions for collecting them were forwarded to the company supplying gas in the particular city from which the samples were desired. At least two 300-c. c. sample bottles were forwarded in each case.

The persons who were to collect the samples were advised to attach a rubber tube to a pipe supplying gas, to invert a container, and to insert the other end of the tube in its mouth. In filling one bottle the gas was to be allowed to flow in five minutes to displace the air originally present, and in filling the other the gas was to flow 10 minutes. The collector was directed to keep the free end of the tube moving about the bottom of the bottle, and on withdrawing the tube, to insert the stopper quickly, keeping the gas directed against the bottle neck meanwhile. Next, melted paraffin was to be poured

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around the stopper and the bottles were to be well packed in wooden cases and mailed to the Pittsburgh laboratory of the Bureau of Mines.

As many of the 5-minute samples contained air, they were discarded and only analyses of 10-minute samples are presented in this report. A few 10-minute samples contained more than 0.1 per cent of oxygen; the analyses of these were rejected and new samples, collected with greater care, were used. When this resampling was done none of the samples finally submitted contained more than 0.1 per cent of oxygen.

METHODS OF EXAMINATION.

The methods of analysis were essentially the same as those described in Bulletin 42 of the Bureau of Mines, "The Sampling and Examination of Mine Gases and Natural Gas." Carbon dioxide was removed with caustic potash solution and oxygen with alkaline pyrogallate solution. The paraffin hydrocarbons were determined by burning them in oxygen and calculating the carbon dioxide and contraction produced to methane or to methane and ethane. Nitrogen was determined by difference. No hydrogen, olefine hydrocarbons, carbon monoxide or hydrogen sulphide was found by qualitative tests. The burette attached to the gas-analysis apparatus with which the analyses were made could not be read much closer than 0.1 c.c. hence a contraction in volume of less than 0.10 c.c. was not recorded.

SPECIFIC GRAVITY.

The specific gravity was generally determined by calculation from the analysis but in some cases by means of the effusion method first proposed by Bunsen.

HEATING VALUE.

The heating values were determined by calculation and are gross heating values. Thomsen's [a] values were used, these being as follows for 0° C. and 760 mm.: Methane (CH_4), 1,065.0 B. t. u.; ethane (C_2H_6), 1,861.0 B. t. u.

TABULAR STATEMENT OF RESULTS.

Essential information regarding the samples is given in the table following.

[a] Landolt-Bornstein, Physikalisch Chemische Tabellen, vol. 5, 1905, p. 425.

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TABLE I.—*Results of examination of samples of natural gas from 25 cities.*

Date of sampling.	Laboratory No.	State.	County.	Town.	C_nH_{2n+2}	CH_4	C_2H_6	CO_2	N_2	O_2	B. t. u. per cubic foot (760 mm. pressure).		Specific gravity.	
											0°C.	60°F.	Calculated.	Determined.
1914.					*P. ct.*	*P. ct.*	*P. ct.*	*P. ct.*	*P. ct.*	*P. ct.*				
November 9	5605	Arkansas and Texas.	Miller and Bowie	Texarkana	96.0	96.0	0.0	0.8	3.2	0.0	1,022	967	0.58	.....
May 8	5275	Indiana	Hamilton	Noblesville	93.0	86.8	6.2	0.8	6.2	0.0	1,040	984	0.62	.....
September 8	5113	Kansas	Allen	Iola	97.7	96.4	1.3	0.9	1.4	0.0	1,051	994	0.58	0.58
September	5351	do	Shawnee	Topeka	95.5	88.8	6.7	0.8	3.7	0.0	1,070	1,013	0.61	.....
October	5105	do	Leavenworth	Leavenworth	95.8	91.3	4.5	0.8	3.4	0.0	1,066	999	0.60	.....
August	5590	Kentucky	Jefferson	Louisville	98.2	77.8	20.4	0.0	1.8	0.0	1,205	1,143	0.66	.....
November 21	5344	Louisiana	Caddo	Lewis	96.5	96.5	0.0	1.4	2.1	0.0	1,028	972	0.58	.....
Do	5643	do	De Soto	Mansfield	97.3	97.3	0.0	0.4	2.3	0.0	1,066	1,009	0.59	.....
November 6	5588	Missouri	Jasper	Joplin	96.9	92.6	4.3	0.6	2.5	0.0	1,066	1,009	0.59	.....
July 7	5271	New York	Allegany	Alma	99.9	68.8	31.1	0.0	0.1	0.0	1,312	1,241	0.71	.....
April	5277	do	Erie	Buffalo	95.1	79.9	15.2	0.0	4.9	0.0	1,134	1,073	0.65	.....
March	5269	Ohio	Hamilton	Cincinnati	99.3	79.8	19.5	0.0	0.7	0.0	1,213	1,147	0.65	.....
July	5101	do	Cuyahoga	Cleveland	98.7	80.5	18.2	0.0	1.3	0.0	1,196	1,131	0.65	.....
Do	5198	do	Washington	Marietta	98.8	73.2	25.6	0.0	1.2	0.0	1,256	1,188	0.69	.....
July 10	5273	do	Clark	Springfield	95.0	80.3	14.7	0.0	5.0	0.0	1,129	1,068	0.65	.....
February	4982	do	Franklin	Columbus[a]	98.5	80.4	18.1	0.0	1.5	0.0	1,193	1,129	0.65	.....
August 27	5403	Oklahoma	Washington	Bartlesville	95.5	92.4	3.1	1.4	3.1	0.0	1,042	986	0.60	.....
August	5373	do	Rogers	Chelsea	93.1	75.4	17.7	0.3	6.6	0.0	1,132	1,071	0.67	.....
August 8	5101	do	Logan	Guthrie	90.0	69.4	20.6	0.1	9.9	0.0	1,122	1,062	0.70	.....
October	5393	do	Muskogee	Muskogee	96.2	92.1	4.1	0.4	3.4	0.0	1,057	1,000	0.59	.....
September	5307	do	Nowata	Nowata	95.2	95.2	0.0	1.3	3.5	0.0	1,014	960	0.58	.....
Do	5415	do	Osage	Pawhuska	94.2	88.6	5.6	1.4	4.4	0.0	1,018	991	0.61	0.64
September 6	5418	do	do	do	87.2	66.5	20.7	0.3	12.5	0.0	1,093	1,034	0.71	0.72
August	5387	do	do	do	93.9	85.1	8.8	1.0	5.1	0.0	1,070	1,012	0.63	0.65
Do	5324	Pennsylvania	Clarion	Oil City	98.9	67.6	31.3	0.0	1.1	0.0	1,302	1,232	0.71	.....
Do	5312	Texas	Navarre	Corsicanna	98.0	98.0	0.0	0.7	1.3	0.0	1,044	987	0.57	.....
October	5445	do	Dallas	Dallas	61.5	50.6	10.9	0.1	38.4	0.0	742	702	0.77	0.76
Do	5443	do	do	do	61.7	51.3	10.4	0.1	38.2	0.0	740	700	0.76	0.76
Do	5345	do	Tarrant	Fort Worth	61.7	51.5	10.2	0.0	38.3	0.0	738	698	0.76	.....
September	5375	do	do	do	61.1	51.1	10.0	0.0	38.9	0.0	730	691	0.76	.....
August	5477	do	do	do	61.5	50.6	10.9	0.1	38.4	0.0	742	702	0.77	.....
Do	5479	do	do	do	61.3	50.6	10.7	0.1	38.6	0.0	738	698	0.77	0.77

[a] Sample collected at Lord Hall, Ohio State University.

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COMMENTS ON THE SAMPLES.

The tabulated results are self-explanatory and need little comment. Five of the samples contain methane only as the combustible constituent. The others also contain higher members of the paraffin hydrocarbon series, principally ethane. Although only methane and ethane are reported, propane is undoubtedly present as well as the butanes and traces of the vapors of the liquid paraffin hydrocarbons, such as pentane and hexane.

In making the ordinary calculation from the combustion data in the analysis, only the two predominating paraffins are shown. The total paraffins are shown correctly, as well as the calculated heating value and specific gravity. The total paraffin content is based on the following equations:

Methane, ethane, propane, and butane react with oxygen as follows:

$$CH_4 \text{ (methane)} + 2O_2 = CO_2 + 2H_2O$$
$$C_2H_6 \text{ (ethane)} + 3.5O_2 = 2CO_2 + 3H_2O$$
$$C_3H_8 \text{ (propane)} + 5O_2 = 3CO_2 + 4H_2O$$
$$C_4H_{10} \text{ (butane)} + 6.5O_2 = 4CO_2 + 5H_2O$$

Let it be assumed that in 100 volumes of these paraffins there are 80 volumes of CH_4, 15 volumes of C_2H_6, 4 volumes of C_3H_8, and 1 volume of C_4H_{10}.

This mixture with sufficient oxygen for complete combustion will produce 126 volumes of CO_2 and contract 213 volumes. The combustion data, if calculated as methane and ethane, are equivalent to 74 volumes of methane and 26 volumes of ethane. The proportions are incorrect, but the amount of total paraffins is exact. The burning of 74 volumes of methane and 26 volumes of ethane consume the same amount of oxygen as the 100 volumes of the mixture.

PROPORTIONS OF PARAFFIN HYDROCARBONS.

In order to determine definitely the quantity of the individual paraffin hydrocarbons that may be present, the Bureau of Mines subjected the natural gas of Pittsburgh to fractional distillation in a vacuum at low temperatures.[a] It is probable that all natural gas of approximately the composition of that used in Pittsburgh contains approximately the same relative proportions of the different paraffin hydrocarbons. The results and those of an ordinary combustion analysis are given below:

Paraffin hydrocarbons in natural gas used in Pittsburgh.

	By fractionation analysis, per cent.	By combustion, per cent.
Nitrogen	1.6	1.2
Methane	84.7	79.2
Ethane	9.4	19.6
Propane	3.0	.....
Butanes (chiefly)	1.3	.....
	100.0	100.0

[a] Burrell, G. A., Seibert, F. M., and Robertson, I. W.: Analyses of natural gas and illuminating gas by fractional distillation in a vacuum at low temperatures; Tech. Paper 104, Bureau of mines. (In press.)

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The predominance of methane and ethane is probably true of all natural gas containing higher members of the paraffin series. The bureau has no data at hand to show that only two paraffins—say methane and ethane—may be present in natural gas, and the presence of ethane or ethane and propane only seems improbable. For comparison with the fractionation analysis of the natural gas of Pittsburgh, given above, there is shown below a fractionation analysis of a so-called "wet gas" from which gasoline is condensed on a commercial scale.

Fractionation analysis of natural gas from which gasoline is condensed.

	Per cent.
Nitrogen	
Methane	36.8
Ethane	32.8
Propane	21.1
Butane (chiefly)	5.8
Pentane and hexane	3.5
	100.0

There is about 1 per cent of nitrogen in the gas, but the nitrogen is not shown in the above analysis.

Considerable difference will be noted between the quantity of the different paraffins in this gas mixture and the one first described. The natural gas of Pittsburgh is typical of gas used in immense quantities in the East for domestic and other purposes. The other represents a gas used for the condensation of gasoline.

COMPARISON OF SAMPLES OF NATURAL GAS FROM DIFFERENT SANDS IN SAME LOCALITY.

The authors have found that in one gas sand methane only may be the combustible constituent of the gas and in another sand a few hundred feet deeper the gas may contain other members of the paraffin series, as is shown by the two analyses following:

Analyses of natural gas from Murrysville and from Elizabeth sands near Trafford City, Westmoreland County, Pa.

Constituents.	Gas from Murrysville sand, depth 1,700 feet.	Gas from Elizabeth sand, depth 2,295 feet.
	Per cent.	*Per cent.*
CO_2	Trace.	Trace.
CH_4	98.8	94.0
C_2H_6	.....	5.2
N_2	1.2	.8
	100.0	100.0

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If a common origin for the natural gas in the two sands be assumed, then the sand in the upper strata has been freed of its ethane and higher paraffins by some method of separation.

CHLORINATION OF NATURAL GAS.

It seems likely that the natural gas high in methane may be of value for the preparation of substitution products, such as chloroform and carbon tetrachloride. Methane can be chlorinated, of course, but the reaction is so slow that it is not in commercial use.

CONDENSATION OF GASOLINE FROM NATURAL GAS.

Those gases containing the higher paraffin hydrocarbons are, of course, of greater heating value and some of them may be used for the extraction of gasoline. About 24,000,000 gallons of gasoline was made from such natural gas in 1913. All of the samples listed in Table 1 do not appear suitable for gasoline extraction.

RELATIVE HEATING VALUE OF DIFFERENT NATURAL-GAS SAMPLES.

Many of the samples in the table show approximately the same heating value. Attention should be called, however, to those used in Dallas and Fort Worth, Tex. (Nos. 5445, 5443, 5345, 5375, 5477, and 5479), with a heating value of about 740 B. t. u. per cubic foot at 0° C. (32° F.) and 760 millimeters pressure. They are exceptional. At the other extreme is the gas used in Alma, N. Y. (No. 5271), with a heating value of 1,312 B. t. u. The samples from gas used in Columbus, Cleveland, Pittsburgh, Cincinnati, and Louisville rank the highest, and in general natural gas from the Appalachian region has the highest heating value.

EXPLOSIBILITY OF MIXTURES OF NATURAL GAS AND AIR.

The Bureau of Mines has determined the explosive limits of mixtures of air and the natural gas used in Pittsburgh, this gas being typical of that supplied to many cities. For most practical purposes these limits may be accepted as applying quite closely to practically all the natural gas mentioned in this paper.

The limits were determined in a Hempel explosion pipette over mercury. Ignition was brought about from above by means of a small electric spark from an induction coil, driven by four dry cells. The smallest quantity of natural gas in a mixture of natural gas and air that upon ignition completely inflamed as observed by the eye was 4.92 per cent. The largest proportion was 11.50 per cent. In its determinations of explosive limits of methane-air mixtures the Bureau of Mines has found that large containers (130 liters capacity) and ignition from below with a large electric flash give

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wider limits than a small vessel and ignition from the top. In the case of methane-air mixtures the difference on the low limit was 0.5 per cent, this is probably true of natural gas-air mixtures; in other words, instead of 4.92 per cent natural gas being the low limit, under other conditions the limit may be as low as 4.50 per cent. The upper limit is undoubtedly extended as much.

AMOUNT OF AIR REQUIRED FOR COMPLETE COMBUSTION.

For most of the samples listed in the table the amount of air required for complete combustion varies but little. One cubic foot of pure methane requires 9.5 cubic feet of air; 1 cubic foot of the natural gas used in Pittsburgh requires 10 cubic feet of air.

AIR OR OXYGEN IN NATURAL GAS.

The authors of this publication could not detect oxygen in the samples of natural gas mentioned in this report (that is, within the error of burette readings, 0.10 per cent). It is their opinion that oxygen, when present (in appreciable quantities) in natural-gas samples is in most cases from air that leaked in, owing to faulty methods of sampling, while the sample was being collected, or at some stage of its transportation or analysis. That natural gas used in cities may be free of air for years has been shown by the authors' experience in analyzing the natural gas used in Pittsburgh. This gas is used at the Pittsburgh station of the Bureau of Mines for different purposes and has been tested regularly for five years. At no time has air been detected in it.

CARBON MONOXIDE, HYDROGEN, AND OLEFIN HYDROCARBONS IN NATURAL GAS.

No carbon monoxide, hydrogen, and olefin hydrocarbons were found in any of the samples of natural gas listed herein, although all of them were carefully tested for these constituents. The authors believe that these constitutents are not present in natural gas, although many published analyses contain them. A careful study of the sources of error that enter into the analyses of natural gas previously made by the Bureau of Mines [a] showed that in the ordinary methods of analyses as applied to natural gas, unavoidable errors creep in. These are principally due to the fact that higher members of the paraffin hydrocarbons are slightly soluble in some of the reagents ordinarily used in gas analyses, such as fuming sulphuric acid, cuprous-chloride solution, and bromine water, and also to the fact that some analysts used such a small proportion of natural gas

[a] Burrell, G. A., and Seibert, F. M., The sampling and examination of mine gases and natural gas: Bulletin 42, Bureau of Mines, 1913, 116 pp.

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for the combustion analyses that a small experimental error was multiplied many times when the percentage calculation was made. The Bureau of Mines of Ontario recently published the analyses [a] of 41 samples of natural gas collected in Ontario. Special tests were made for carbon monoxide, hydrogen, and olefin hydrocarbons, and they were not detected. Neither was oxygen detected in any of the samples except in traces.

The same can be said of the much earlier work of Phillips,[b] and also of the recent work of Czako.[c] Cady and McFarland,[d] however, report small quantities of carbon monoxide and olefine hydrocarbons in many samples they examined. A study of their methods of examination reveals that the methods they used in testing for these constitutents were at fault, in that as a particular sample was passed into the absorbents in making an analysis, some of the paraffin hydrocarbons were removed and not, as they thought, carbon monoxide or olefins. Many other analyses, too numerous to mention here, report these constituents. Without doubt they must be regarded with suspicion; invariably they were analyzed without necessary extra precaution and the consequent errors could not be avoided.

IGNITION TEMPERATURE OF NATURAL GAS.

To the authors' knowledge no direct determination of the ignition temperature of natural gas has been made. The ignition temperature [e] of methane in air is 650° C. to 750° C. and of ethane in air is 520° C. to 630° C. From this one would conclude that the ignition temperature of most natural gas, perhaps all of it, is close to 550° C. to 750° C.

THEORETICAL PROPORTIONS OF NATURAL GAS AND AIR.

The bureau determined by experiment that the mixture of air and the natural gas of Pittsburgh, containing the maximum proportion of natural gas that will burn with complete combustion of the air and gas is 8.6 per cent. Many of the samples of natural gas listed herein would run about the same.

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[a] Mickle, G. R., The chemical composition of natural gas found in Ontario (Chemical analyses by W. H. Ellis, J. W. Bain, and E. G. R. Ardagh): 23d Rept. Bureau of Mines of Ontario, 1914, p. 13.

[b] Phillips, F. C., Petroleum and natural gas precise levels: W. Va. Geol. Survey, vol. 1-A, 1904, p. 513; Researches upon the phenomena of oxidation and chemical properties of gases: Am. Chem. Jour., vol. 16, 1894, p. 406.

[c] Czako, Emerich, Contribution to the knowledge of natural-gas outbursts. Dissertation. Technical High School of Karlsruhe. 1913. 85 pp.

[d] Cady, H. P., and McFarland, D. F., The occurrence of helium in natural gas and the composition of natural gas: Jour. Am. Chem. Soc., vol. 29, 1907, p. 1523.

[e] Dixon, H. B., and Coward, H. F., The ignition temperatures of gases: Chemical News, vol. 99, 1909, p. 139.

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CONSTANT COMPOSITION OF THE NATURAL GAS OF PITTSBURGH.

The following analyses of the natural gas of Pittsburgh were made for a period of almost one year. It will be noted that during that time the gas remained remarkably constant in composition. The many different wells from which the gas is obtained must contain gas of very nearly the same composition.

Composition of Pittsburgh natural gas.

Test No.	Date.	C_nH_{2n+2}.	CH_4.	C_2H_6.	CO_2.	O_2.	N_2.	B. t. u. per cubic foot at 760 mm. pressure.		Specific gravity, calculated.
								0° C.	60° F.	
	1912.	*P. ct.*	*P. ct.*	*P. ct.*	*P. ct.*	*P. ct.*	*P. ct.*			
1	Dec. 7	99.3	80.8	18.5	0.0	0.0	0.7	1,205	1,140	0.65
	1913.									
2	Apr. 7	99.3	80.4	18.9	.0	.0	.7	1,208	1,143	.65
3	Apr. 21	99.2	80.7	18.5	.0	.0	.8	1,205	1,139	.65
4	May 9	99.1	78.7	20.4	.0	.0	.9	1,218	1,152	.66
5	June 2	99.0	81.7	17.3	.0	.0	1.0	1,192	1,128	.64
6	Nov. 8	99.2	83.4	15.8	.0	.0	.8	1,182	1,119	.64
7	Dec. 13	99.3	81.7	17.6	.0	.0	.7	1,198	1,133	.64
	1914.									
8	Jan. 2	99.2	82.0	17.2	.0	.0	.8	1,193	1,129	.64
9	Jan. 31	99.3	80.9	18.4	.0	.0	.7	1,204	1,139	.65
10	Feb. 16	98.9	81.0	17.9	.0	.0	1.1	1,196	1,131	.65
11	May 1	99.4	81.0	18.4	.0	.0	.6	1,205	1,140	.65
12	June 16	99.4	80.9	18.5	.0	.0	.6	1,206	1,141	.65
13	July 1	98.9	78.9	20.0	.0	.0	1.1	1,212	1,147	.66
14	Aug. 16	99.1	80.7	18.4	.0	.0	.9	1,202	1,137	.65
15	Sept. 1	99.1	80.4	18.7	.0	.0	.9	1,204	1,139	.65
16	Sept. 20	99.3	79.3	20.0	.0	.0	.7	1,217	1,151	.66
17	Sept. 31	99.1	79.0	20.1	.0	.0	.9	1,215	1,150	.66
18	Oct. 16	99.4	79.5	19.9	.0	.0	.6	1,217	1,151	.66
19	Nov. 2	99.4	80.7	18.7	.0	.0	.6	1,209	1,142	.65
20	Nov. 17	99.2	80.5	18.7	.0	.0	.8	1,205	1,140	.65

FLUCTUATION IN COMPOSITION OF OTHER NATURAL GAS.

Of much interest in its bearing on the fluctuation in composition of natural gas is the work of Phillips.[a] Phillips determined the fluctuation in the composition by measuring the nitrogen. The following table shows the variation in nitrogen content as found by him for gas from different wells.

Fluctuation in nitrogen percentage of natural gas.

Well.	Variation in percentage of nitrogen.	Number of samples collected.	Period over which samples were collected.
Gas well at Shields, 14 miles west of Pittsburgh, Pa.	1.10 to 2.70	6	Aug. 5, 1896, to June 1, 1898.
Gas well on Anderson farm near Sewickley, Pa.	1.71 to 2.50	3	July 7, 1896, to Mar. 22, 1897.
Gas well on Miller farm at Glenfield, Pa., 9½ miles west of Pittsburgh.	1.22 to 3.25	9	July 27, 1896, to June 13, 1898.
Gas well on Bayley farm, Neville Island, in the Ohio River, 6½ miles west of Pittsburgh.	1.46 to 2.11	4	Aug. 1, 1896, to May 27, 1898.

[a] Phillips, F. C., Fluctuations in the composition of natural gas; and a method for the determination of nitrogen in gas mixtures: Proc. Eng. Soc. Western Pa., vol. 14, December, 1898, pp. 290–307.

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He also examined samples of gas from seven other wells. They all showed about the same results as those mentioned in the table—namely, only a comparatively slight variation in nitrogen content, and hence of other constitutents, although in some cases the tests extended over two years.

RARE GASES IN NATURAL GAS.

The principal inert dilutant of natural gas is nitrogen, the proportion of which varies from 1 per cent or less to 30 or 40 per cent in gases used commercially and in exceptional cases much higher. A sample of marsh gas obtained from the State of Washington contained, according to analyses made by the authors, as much as 98.5 per cent of nitrogen. This sample was taken from a small outburst of gas going to waste.

In ordinary gas analyses the nitrogen content is determined by difference, no attempt being made to isolate the so-called rare gases such as are associated with nitrogen in atmospheric air. The presence of these rare gases in natural gas is at present principally of academic interest, as they are inert like nitrogen. That they are present, however, in small proportions has been shown by several investigators. Cady and McFarland [a] determined that helium was present in 41 samples, obtained from Kansas and other States, in quantities varying from a trace up to 1.84 per cent. The helium tended to increase with the nitrogen but showed no direct relation.

Czako [b] also found small quantities of helium in 6 samples of natural gas he investigated, in proportions ranging from 0.0010 per cent to 0.41 per cent. Other investigators have found the inactive rare gases in earth gases, in connection with work on radioactive emanations. Helium especially has been the subject of considerable research, as it is regarded as the final product of all radioactive decomposition processes.

Numerous investigations begun about 10 years ago showed that the majority of gas outbursts from the earth carry with them a radioactive emanation, which they have absorbed while passing through the earth strata within which the radioactive process of decomposition takes place. In many cases the emanation proved to be radium. Numerous minerals, rocks, soils, underground air and atmospheric air, as well as water and gases from springs, have been examined for their radioactivity, often in connection with their helium content.

[a] Cady, H. P., and McFarland, D. F., The occurrence of helium in natural gas and the composition of natural gas: Jour. Am. Chem. Soc., vol. 29, 1907, p. 1523.

[b] Czako, Emerich, Contribution to the knowledge of natural-gas outbursts. Dissertation. Technical High School Karlsruhe. 1913. 85 pp.

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PHYSIOLOGICAL EFFECTS OF NATURAL GAS.

EFFECTS OF NATURAL GAS ON CANARIES.

The artificial illuminating gas of cities, made by the destructive distillation of coal and oil or by the action of steam on hot coke, contains from 8 or 10 per cent upward of carbon monoxide. In some cities the gas may contain as much as 30 per cent of carbon monoxide, and many deaths have been caused from its escaping and the user breathing it. On the other hand, natural gas contains no carbon monoxide or other poisonous gases, excepting hydrogen sulphide in occasional samples. But, to the authors' knowledge, natural gas containing hydrogen sulphide is not used in any city. However, there are present in the natural gas used in many cities small proportions of the higher paraffin hydrocarbons, butane and pentane. These if inhaled in sufficient quantity exert an anesthetic action on the human system. It is generally known, for instance, that ordinary gasoline, which contains large quantities of pentane and hexane, will give many people a headache if inhaled in sufficient quantity and often has produced a condition similar to alcoholic intoxication. Methane, the chief constituent of natural gas, is physiologically inert. Perhaps the same can be said of ethane, but most likely propane has some effect and butane and pentane undoubtedly have. In most natural gas the proportions of butane and pentane are probably too small to have any decidedly dangerous effect on man.

In order to get some information on this matter some experiments were made on canaries. Various proportions of the natural gas used in Pittsburgh were introduced with pure air in a 10-liter bell jar and the effects noted. The results were as follows:

Effect on canaries of breathing mixtures of natural gas and air.

Test No.	Natural gas.	Air.		Carbon dioxide.	Effect on canary.
		Oxygen.	Nitrogen.		
	Per cent.	*Per cent.*	*Per cent.*	*Per cent.*	
1	76.9	4.8	18.2	0.1	Immediate collapse.
2	69.9	6.1	23.1	.9	Do.
3	66.3	6.9	26.1	.7	Showed immediate distress, but did not collapse in 1 hour's time.
4	65.8	7.1	26.9	.2	Immediate collapse.
5	63.0	7.5	28.4	1.1	Showed immediate distress, but did not collapse in 1 hour's time.
6	49.3	10.6	40.0	.1	Showed unsteadiness of movement for the first 5 minutes and was more or less drowsy during the exposure, which lasted 1 hour.
7	44.5	11.6	43.8	.1	Do.
8	34.5	13.7	51.7	.1	Only symptom in 1 hour's time was drowsiness and unsteady movement.

According to the foregoing table, the proportion of natural gas in air required to produce collapse in canaries is very large (65 per cent or more). With as much as 63 per cent collapse did not occur in

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one hour's time, although the distress was immediate. Distress was evinced by the canaries opening their bills and breathing rapidly and finally squatting on the perch in a lethargic manner. Some of the canaries were more resistant than others.

COMPARISON WITH EFFECTS OF ATMOSPHERES DEFICIENT IN OXYGEN.

EFFECT ON CANARIES OF ATMOSPHERES LOW IN OXYGEN.

The results of these experiments indicate that as far as the action of natural gas on canaries is concerned it is largely a question of a deficiency of oxygen produced by diluting the air with natural gas. In investigations to determine the suitability of using small animals in mines to detect atmospheres dangerously low in oxygen, the results of which are described in Technical Paper 122,[a] experiments were made to determine the resistance of canaries to low-oxygen atmospheres produced by diluting air with nitrogen. The results obtained, as may be seen from the table following, show that the effect on the canaries is much the same as that of natural gas.

In conducting the experiments, atmospheres containing various percentages of nitrogen and oxygen were prepared in bell jars of 10 liters capacity. The nitrogen used analyzed 97.8 per cent nitrogen and 2.2 per cent oxygen. Three canaries, designated in the table as A, B, and C, were used in the experiments.

Effect on canaries of breathing atmospheres low in oxygen.

Test No.	Canary used.	Composition of atmosphere.					
		At beginning of experiment.			At end of experiment.		
		Oxygen.	Nitrogen.	Carbon dioxide.	Oxygen.	Nitrogen.	Carbon dioxide.
		Per cent.	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*
1	B	9.42	90.48	0.10	8.50	90.90	0.60
2	C	9.25	90.65	.10	.....	.....	.....
3	A	7.65	92.25	.10	7.55	92.05	.40
4	C	7.83	92.07	.10	.....	.....	.....
5	A and C	7.10	92.80	.10	.....	.....	.....

REMARKS.

Test 1.—The canary, B, showed immediate distress as evinced by rapid breathing, open bill, and unsteadiness. In 10 minutes' time the bird was apparently in normal condition, with the exception of slight increase in the rate of breathing. It was removed from the atmosphere at the end of 1 hour's time.

Test 2.—The behavior of the canary, C, was similar to that of the bird in the previous experiment.

Test 3.—The canary, A, showed immediate distress by panting and unsteadiness, but did not collapse or evince any more distress than the bird in test No. 1. It was left in the atmosphere for 1 hour.

[a] Burrell, G. A., and Oberfell, G. G., Relative effects of atmospheres deficient in oxygen on small animals and on men: Technical Paper 122, 1915.

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Test 4.—The canary, C, collapsed as soon as it was placed in the atmosphere. It breathed very slowly with eyes and bill closed, but recovered its normal state in less than a minute after it was removed to fresh air.

Test 5.—One canary, A, showed distress but did not collapse in 20 minutes' time. Another canary, C, was placed in the atmosphere and immediately collapsed.

The canaries showed some distress in atmospheres containing 9.4 per cent of oxygen, more pronounced distress in atmospheres containing as low as 7.6 or 7.8 per cent, and in an atmosphere containing as little as 7.1 per cent of oxygen may or may not collapse.

EFFECT ON MEN OF ATMOSPHERES LOW IN OXYGEN.

In a test to determine the effects on man of atmospheres low in oxygen, described in Technical Paper 122, a member of the Bureau of Mines breathed air in and out of a bag having a capacity of about 70.0 liters, the exhaled carbon dioxide being removed by means of a can of caustic potash inserted between his mouth and the bag, and was rendered unconscious when the oxygen content of the air had fallen to about 7.0 per cent. The mode of action of the "oxygen want" is instructive. The subject felt warning symptoms previous to collapse, but did not believe himself in any danger of losing consciousness, and, in fact, wanted to continue the experiment. He felt no real distress until some time after the experiment, but on the next day was decidedly unwell.

In its insidious action "oxygen want" acts as carbon monoxide frequently does, that is, when the oxygen is slowly decreased. One difficulty in comparing the mode of action of the two lies in the scarcity of experimental data on the effects on men of atmospheres low in oxygen.

Some light is thrown on the action of low-oxygen atmospheres by an accident that occurred in the Lodge Mill colliery, Huddersfield, England,[a] in which three men were overcome by black damp in an unused part of the mine. Two men, A and B, were overcome by the black damp at 3.30 a. m. A third man, C, was overcome at 4 a. m. in an attempt to rescue his comrades. At 1 p. m. of the same day rescuers equipped with breathing apparatus removed C, and about an hour later removed A. A little later B was found dead. Both C and A were in a critical condition, but still breathing. A died three days later. The air in which the men were overcome was high in methane and correspondingly low in oxygen. Haldane[b] comments on this disaster as follows:

All the facts recorded indicate that the men were overcome by the insufficiency in the oxygen percentage of the air. There was no reason to suspect the presence of carbon monoxide as there was no gob fire or heating. In addition, the blood of the dead

[a] See Lloyd, W. D., The use of rescue apparatus at Lodge Mill colliery, Huddersfield; with a note by J. S. Haldane: Coll. Guard., Nov. 7, 1913, vol. 106, pp. 957–958.

[b] Haldane, J. S., Note on paper by W. D. Lloyd: Coll. Guard., vol. 106, 1913, p. 958.

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man was black, not red, as it would have been if death was due to carbon monoxide. It is probable that sufficient fire damp was present to reduce the oxygen to 7 or 8 per cent. The fact that A did not recover was due to exactly the same cause which often prevents men recovering after severe carbon-monoxide poisoning. The tissues have been severely damaged by the prolonged exposure to dearth of oxygen, so that although the oxygen supply is completely restored recovery is doubtful. In the case of A the post-mortem examination revealed the fact that the heart was dilated. Probably the heart muscles and the other tissues were in a condition of fatty degeneration caused by the want of oxygen. The writer has seen other similar cases of dilation simulating severe heart disease, and only slowly recovering, after prolonged exposure to carbon-monoxide poisoning. So far as he is aware, however, this is the only recorded case of death after partial recovery from exposure to an atmosphere which was simply deficient in oxygen, apart from the presence of carbon monoxide.

In regard to the oxygen deficiency required to cause distress in men Haldane [a] says:

When the oxygen content of the air is gradually reduced by the absorption of the oxygen or (what is exactly the same thing) by addition of nitrogen, very little may be felt before the occurrence of impairment of the senses and loss of power over the limbs. If the reduction is gradual and the symptoms be carefully watched it will be noticed that at about 12 per cent oxygen—that is with a reduction of 9 per cent—the respirations become just perceptibly deeper. At 10 per cent the respirations are distinctly deeper and more frequent, and the lips become slightly bluish. At 8 per cent the face begins to assume a leaden color, though the distress is still not great. With 5 or 6 per cent there is marked panting, and this is accompanied by clouding of the senses and loss of power over the limbs, which would probably end sooner or later in death. It is probable that any sudden exertion made in air markedly deficient in oxygen may lead to temporary loss of consciousness, so that sudden efforts should be avoided in all cases where, through accident or necessity, a man is in an atmosphere which will not support light and in such a position that he might fall into worse air or otherwise injure himself. When air containing less than 1 or 2 per cent of oxygen is breathed, loss of consciousness, without any distinct warning symptoms, occurs within 40 or 50 seconds. Loss of consciousness in air deprived of oxygen is more rapid than in drowning or strangling, since in the former case not only is the supply of fresh oxygen cut off but the oxygen previously in the lungs is rapidly washed out, loss of consciousness is quickly followed by convulsions, which are followed by cessation of the respiration. The heart still continues to beat, in the case of cats and dogs, for from 2 to 8 minutes; in men this period is probably much longer, for it seems to be the general rule that the larger the animal the longer it resists asphyxiation. So long as the heart is beating, however feebly, animation may be restored by artificial respiration. This may require to be continued for a considerable period, as the after effects of deprivation of oxygen are very serious and the respiratory center may not recover for some time.

The experiments cited show that oxygen deprivation begins to affect men in about the same time it does canaries, and the percentages that cause collapse are not much different. Atmospheres that will cause the animals to collapse vary to a certain extent, and presumably the same is true of men.

[a] Haldane, J. S., The causes of deaths in colliery explosions and underground fires: Report to the Secretary of State for the Home Dept., 1896, p. 15.

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EFFECT OF NATURAL GAS ON MAN.

As far as the diluting action of nitrogen and natural gas in lowering the oxygen content of air is concerned, the two are nearly similar in the effects produced on canaries. Possibly the same is true as regards man, but to make certain of this point experiments would have to be made on human subjects. One is safe, however, in drawing the conclusion that a large proportion of natural gas in mixture with air is required to affect man and that the danger of death to the occupants of a room because of natural gas escaping from an open gas jet is remote, because of the very large quantity of gas required. An ordinary natural-gas burner in a house, used for giving light, burns about 4 cubic feet per hour. At this rate, to reduce the proportion of oxygen in the air of a room 9 feet high and 15 feet square to 10 per cent, the burner would have to flow gas for about 263 hours, assuming that no gas leaked out of the room through the door jambs, window jambs, and cracks in the walls (an impossibly tight condition, by the way). Therefore the possibility of suffocation being produced by natural gas under such conditions is very remote. Of course, in a small room where several burners were flowing the danger would be greater. The danger of explosion from the accidental lighting of a mixture of natural gas and air is much greater than that of suffocation from it. The proportion of natural gas in air required to make the mixture explosive is about 5 to 12 per cent, whereas to produce suffocation apparently requires about 60 per cent.

Artificial illuminating gas made from coal, on the other hand, may be poisonous when as little as 1 per cent (based on only 10 per cent of carbon monoxide in the gas) of it is in the room, and dangerously so when 2 per cent is present. It is also explosive in relatively small proportion, the low limit being about 7 or 9 per cent and the high limit 18 to 21 per cent.

IMPERFECT COMBUSTION OF NATURAL GAS IN STOVES.

No account is taken in the foregoing of the imperfect combustion of gas, either natural or artificial, in poorly constructed gas stoves. The danger in such cases from either gas is great, and not only should stoves be well constructed, so that perfect combustion takes place, but every stove should be equipped with a vent leading to the house chimney to take away the products of combustion. Although the results to the occupants of the house if this is not done may not be apparent, in that distress is not produced, yet bad effects may develop, due principally to generation of small proportions of carbon monoxide. The public is kept more or less aware of the fatalities produced by the imperfect combustion of gas in stoves by frequent citations of such accidents in the daily newspapers.

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REMOVAL OF PRODUCTS OF COMBUSTION OF NATURAL GAS FROM ROOMS.

Natural gas, when burned with sufficient oxygen for complete combustion, forms carbon dioxide and water vapor. Each cubic foot of natural gas burned produces a little over 1 cubic foot of carbon dioxide and a little more than 2 cubic feet of water vapor. Carbon dioxide is an irrespirable gas and should not be allowed to accumulate in a room. Water vapor also should be removed, because it has a depressing effect if present in still, warm air in sufficient proportion and tends to make the walls, ceilings, curtains, and other objects in a room dirty because the dust is entrained by it and settles on the objects.

The only way to remove these two gases is by means of a vent leading from the stove to the house chimney. It is absurd for any manufacturer of stoves to claim that these two gases are practically absorbed or eliminated in any other way.

SUMMARY.

Out of the samples of natural gas from 25 cities in the United States, 5 contain methane only as the combustible gas. The others contain, in addition to methane, higher members of the series of paraffin hydrocarbons.

Natural gas in two different sands of the same field may vary decidedly in composition.

Some of the natural gas used in Texas has a heating value of about 740 B. t. u. per cubic foot at 0° C, and 760 mm. pressure. The highest heating value determined was 1,312 B. t. u. per cubic foot.

The natural gas used in Pittsburgh, Columbus, Cleveland, Cincinnatti, and many other places in the East is remarkably uniform in composition.

The explosive limits of mixtures of natural gas and air lie between about 5 per cent gas, low limit, and 11.50 per cent gas, high limit. Many of the samples mentioned herein require about 10 cubic feet of air per cubic foot of gas for complete combustion.

According to Cady and McFarland and to Czako, helium may be present in natural gases from a trace to 1.84 per cent.

The ignition temperature of natural gas lies between about 550° C. and 750° C.

The composition of the natural gas used in any one town may remain remarkably constant for a long period.

Oxygen, carbon monoxide, hydrogen, and olefin hydrocarbons are not present in natural gas.

The only practical way to remove products of combustion (either perfect or imperfect) from a gas stove is by means of a vent leading to the house chimney.

A large proportion of natural gas in air is required to suffocate men. As far as canaries are concerned, the effect on them of mixtures of natural gas and air is principally due to the lowering of the oxygen content of the air by the diluting action of the gas.

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BULLETIN 88. The condensation of gasoline from natural gas, by G. A. Burrell, F. M. Seibert, and G. G. Oberfell. 1915. 106 pp., 6 pls., 18 figs.

TECHNICAL PAPER 3. Specifications for the purchase of fuel oil for the Government, with directions for sampling oil and natural gas, by I. C. Allen. 1911. 13 pp.

TECHNICAL PAPER 10. Liquefied products from natural gas; their properties and uses, by I. C. Allen and G. A. Burrell. 1912. 23 pp.

TECHNICAL PAPER 25. Methods for the determination of water in petroleum and its products, by I. C. Allen and W. A. Jacobs. 1912. 13 pp., 2 figs.

TECHNICAL PAPER 32. The cementing process of excluding water from oil wells as practiced in California, by Ralph Arnold and V. R. Garfias. 1913. 12 pp., 1 fig.

TECHNICAL PAPER 37. Heavy oil as fuel for internal-combustion engines, by I. C. Allen. 1913. 36 pp.

TECHNICAL PAPER 38. Wastes in the production and utilization of natural gas and means for their prevention, by Ralph Arnold and F. G. Clapp. 1913. 29 pp.

TECHNICAL PAPER 42. The prevention of waste of oil and gas from flowing wells in California, with a discussion of special methods used by J. A. Pollard, by Ralph Arnold and V. R. Garfias. 1913. 15 pp., 2 pls., 4 figs.

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TECHNICAL PAPER 74. Physical and chemical properties of the petroleums of California, by I. C. Allen, W. A. Jacobs, A. S. Crossfield, and R. R. Matthews. 1914. 38 pp., 1 fig.

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TECHNICAL PAPER 115. Inflammability of mixtures of gasoline vapor and air, by G. A Burrell and H. T. Boyd. 1915.

TECHNICAL PAPER 119. The limits of inflammability of mixtures of methane and air, by G. A. Burrell and G. G. Oberfell. 1915.

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